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It is known that in a certain case, the secondary Bjerknes force, which is a radiation force acting between pulsating bubbles, changes (e.g., from attractive to repulsive) as the bubbles approach each other. In this letter, a theoretical discussion of this phenomenon is given. The present theory, based on analysis of the eigenfrequencies of interacting bubbles, gives an accurate interpretation, which differs from previous ones (e.g., by Doinikov and Zavtrak [Phys. Fluids **7**, 1923-1930 (1995)]), of the phenomenon. It is shown, for example, that the first reversal, occurring when both bubbles are larger than the resonance size and the damping effect is sufficiently small, is due to the highest eigenfrequency of the larger bubble.

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I. INTRODUCTION

It is known that two gas bubbles pulsating in an acoustic field experience an interaction force called the secondary Bjerknes force.¹⁻⁴ In 1984, Zabolotskaya showed theoretically that in a certain case, the sign of the force may change as bubbles come closer to one another.² She assumed that the reversal of the sign is due to the variation in the natural frequencies of the interacting bubbles. However, the theoretical formula derived for evaluating the natural frequencies of two interacting bubbles cannot explain the reversal. The formula is represented as

$$(\omega_{10}^2 - \omega^2)(\omega_{20}^2 - \omega^2) - \frac{R_{10}R_{20}}{D^2}\omega^4 \approx 0 \quad (1)$$

where R_{10} and R_{20} are the equilibrium radii of bubbles, ω_{10} and ω_{20} are their partial natural frequencies, ω is the driving frequency of the external sound, and D is the distance between the centers of the bubbles. This equation predicts the existence of two natural frequencies, and is symmetric; namely, it exchanges 10 and 20 on the subscripts of variables to reproduce the same equation. This means that the two bubbles have the same natural frequencies, and their respective pulsation phases are simultaneously inverted (e.g., from in-phase to out-of-phase with the external sound) at the distance where the (effective) natural frequencies of the bubbles are equal to the driving frequency. As a result, the bubbles pulsating in-phase with each other will sustain in-phase pulsation even when the distance between them becomes sufficiently small.

In 1995, Doinikov and Zavtrak, using a mathematical model in which the multiple scattering of sound between bubbles is taken into account more accurately, predicted again the reversal of the sign.³ They also asserted that the reversal is due to the change in the natural frequencies. They assumed that the natural frequencies of both

bubbles increase as the bubbles approach each other, resulting sometimes in the reversal of the sign. When, for example, both bubbles are larger than the resonance size and the distance between them is large enough, they pulsate in phase. As the bubbles approach each other, the natural frequency of a smaller bubble may first, at a certain distance, rise above the driving frequency, and, in turn, the bubbles' pulsations become antiphase; the force then changes from attractive to repulsive. On the other hand, when one bubble is larger and the other is smaller than the resonance size and the distance between them is large, they pulsate out of phase and the force is repulsive. As the distance between the bubbles becomes small, the natural frequencies of both bubbles may rise, and when the natural frequency of a larger bubble rises above the driving frequency, the repulsive force may turn into attraction. Although this interpretation can explain the reversal, it is in opposition to the prediction given by Eq. (1), which reveals that the higher natural frequency (converging to the partial natural frequency of a smaller bubble for $D \rightarrow \infty$)^{5,6} becomes higher and the lower one (converging to the partial natural frequency of a larger bubble for $D \rightarrow \infty$) becomes smaller as the bubbles approach each other.

The aim of this letter is to give an alternative interpretation of the reversal of the sign; one that may be more accurate than the previous ones. Recently, having reexamined Zabolotskaya's model, in which a system of differential equations for linear coupled oscillators is employed, we found that a bubble interacting with a neighboring bubble has three "eigenfrequencies", defined as *the driving frequencies for which the phase difference between an external sound and the pulsation of the bubble becomes $\pi/2$* .^{5,6} Among the three eigenfrequencies, the lowest one decreases and remaining two increase as the bubbles approach each other. Only one of them converges to the partial natural frequency of the corresponding bubble for $D \rightarrow \infty$. Namely, the eigenfrequencies

defined as above are asymmetric. Using this theory for the eigenfrequencies, we give a novel explanation for the reversal.

II. THEORIES

In this section, we briefly review previously expounded theories for deriving the eigenfrequencies and estimating the magnitude of the secondary Bjerknes force. Let us assume that the pressure of the external sound, p_{ex} , is written in the form of $p_{\text{ex}} = -P_a \sin \omega t$, and the time-dependent radii of bubbles 1 and 2, R_1 and R_2 , respectively, can be represented as $R_1 = R_{10} + e_1$, $R_2 = R_{20} + e_2$ and $|e_1| \ll R_{10}$, $|e_2| \ll R_{20}$, where P_a is a positive constant, and R_{j0} and e_j are the equilibrium radius and the deviation of the radius, respectively, of bubble j ($j = 1, 2$). From the linearized system for coupled oscillators,^{1,5,6} we can obtain the harmonic steady-state solution for e_1 and e_2 , expressed as

$$\begin{aligned} e_1 &= K_1 \sin(\omega t - \phi_1), \\ e_2 &= K_2 \sin(\omega t - \phi_2), \end{aligned}$$

where the concrete expressions for the amplitude K_1 and the phase difference ϕ_1 are given in Refs. 5 and 6. The eigenfrequencies of bubble j are determined so that ϕ_j becomes $\pi/2$.^{5,6} The resulting formula for deriving the eigenfrequencies is

$$H_1 F + M_2 G = 0 \quad (2)$$

with

$$\begin{aligned} F &= L_1 L_2 - \frac{R_{10} R_{20}}{D^2} \omega^4 - M_1 M_2, \\ G &= L_1 M_2 + L_2 M_1, \quad H_1 = L_2 + \frac{R_{20}}{D} \omega^2, \\ L_1 &= (\omega_{10}^2 - \omega^2), \quad L_2 = (\omega_{20}^2 - \omega^2), \\ M_1 &= \delta_1 \omega, \quad M_2 = \delta_2 \omega, \end{aligned}$$

where δ_1 and δ_2 are determined from the damping characteristic of the system (see the following section). The above formula is for bubble 1; exchanging 1 and 2 in the subscripts of variables in this equation yields the formula for bubble 2.

The secondary Bjerknes force acting between the bubbles is expressed with^{1,7}

$$\mathbf{F} \propto K_1 K_2 \cos(\phi_1 - \phi_2) \frac{\mathbf{r}_2 - \mathbf{r}_1}{\|\mathbf{r}_2 - \mathbf{r}_1\|^3},$$

where \mathbf{r}_1 and \mathbf{r}_2 are the positions of bubbles 1 and 2, respectively, and $\|\mathbf{r}_2 - \mathbf{r}_1\| = D$. The reversal of the sign of this force occurs only when the sign of $\cos(\phi_1 - \phi_2)$ changes, because $K_1 > 0$ and $K_2 > 0$ always.^{5,6}

III. RESULTS AND DISCUSSION

First, we discuss the case of $R_{10} = 3$ mm and $R_{20} = 6$ mm, which corresponds to a case used in Ref. 4. We assume that the bubbles are filled with a gas with a constant specific heat ratio of 1.4, and the surrounding material is water. For the damping coefficient, we adopt that used for radiation loss in Ref. 4 (actually, the thermal loss should not be neglected in the frequency range used here; we neglect it, however, in order to simplify the following discussion), and the value for viscous loss was that employed in our previous paper. Thus, we reset δ_1 and δ_2 in Refs. 5 and 6 as $\delta_1 = \omega^2 R_{10}/c$ and $\delta_2 = \omega^2 R_{20}/c$, where c is the speed of sound in water.

Figures 1(a) and 1(a') show the natural frequencies of the bubbles, ω_1 and ω_2 , calculated by using Eq. (2). In those figures, l denotes the normalized distance defined as $l = D/(R_{10} + R_{20})$. As mentioned previously, three eigenfrequencies appear in the region where l is small, and only one eigenfrequency, converging to the partial resonance frequency of the corresponding bubble, remains when l is sufficiently large. The second highest eigenfrequency of bubble 2 is almost equal to the highest one of bubble 1, and the highest one of bubble 2 is, thus, higher than that of bubble 1. Figures 1(b), 1(b'), and 1(c) show $C_1 = \cos(\phi_1)$, $C_2 = \cos(\phi_2)$, and $E = \cos(\phi_1 - \phi_2)$, respectively, as functions of l . Here the driving frequency is assumed to be $\omega = 1.18 \omega_{10}$. When l is large, both bubbles pulsate out-of-phase with the external sound because $\omega > \omega_{10}$ and $\omega > \omega_{20}$; thus, both C_1 and C_2 are about -1 and the force is attractive (i.e., $E > 0$). When l becomes sufficiently small, C_2 first turns positive, and the force consequently changes from attractive to repulsive. This result is inconsistent with the interpretation given in Refs. 3 and 4, both of which claim that the first reversal in this case is due to the rise in the natural frequency of the smaller bubble.

Now let us consider the dependency of the sign of the force on ω . Figure 2 shows E calculated by using different driving frequencies. For $\omega = 1.05 \omega_{10}$, the sign of the force changes three times. We can see from Fig. 1 that, in this case, the eigenfrequencies of bubbles 1 and 2 become equal to ω once and twice, respectively, during the approach; this may cause the complicated behavior of the sign. In the case of $\omega = 0.95 \omega_{10}$ ($\omega < \omega_{10}$ and $\omega > \omega_{20}$), the sign stays negative through out because the eigenfrequencies of both bubbles never become equal to ω . For $\omega = 0.6 \omega_{10}$, one reversal from repulsive to attractive is observed. In Refs. 3 and 4, to explain the last case, it was assumed that the reversal occurs when the natural frequency of the larger bubble, rising as the bubbles approach each other, exceeds the driving frequency. However, the present result reveals that the reversal in this case is due to the second highest eigenfrequency of the smaller bubble. This result cannot be explained by Zabolotskaya's theory either [Eq. (1)], given that it assumes that the lower natural frequency decreases.

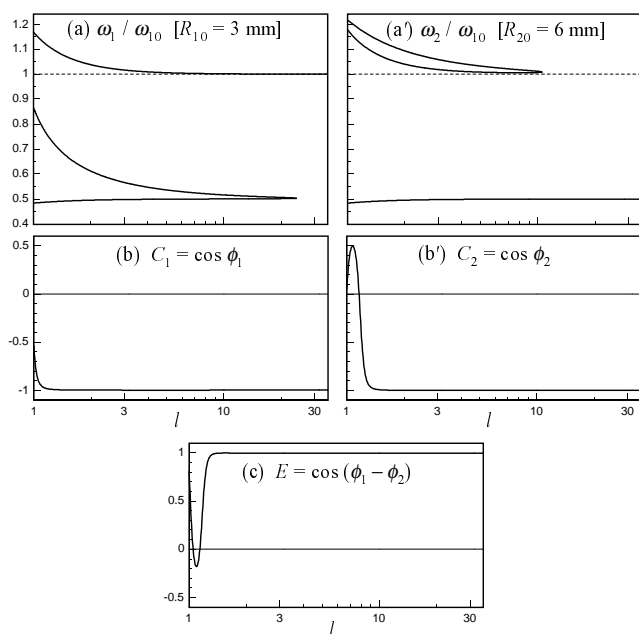


FIG. 1. Eigenfrequencies normalized by ω_{10} [(a) and (a')], pulsations [(b) and (b')], and phase difference between the bubble and the driving frequency is assumed to be $\omega = 1.18\omega_{10}$.

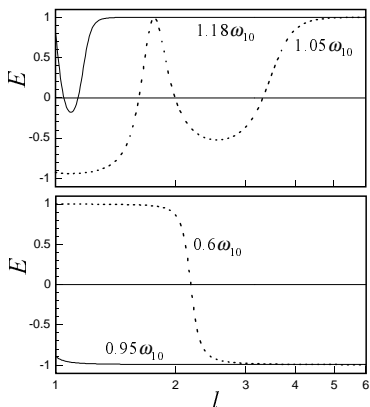


FIG. 2. $E = \cos(\phi_1 - \phi_2)$ for $\omega = 1.18\omega_{10}$, $1.05\omega_{10}$, $0.95\omega_{10}$, and $0.6\omega_{10}$ for convenience of comparison.

Next, we show results for $R_{10} = 20 \mu\text{m}$ and $R_{20} = 30 \mu\text{m}$ (this setting is typical of those used in Ref. 3). In this case, we adapt the thermal loss for the damping factor, as done in Ref. 3. For computing the polytropic exponent and the damping coefficient, we employ Devin's formula and use parameters the same as those used in Ref. 3. Figure 3 shows the eigenfrequencies of both bubbles. In this setting, "subeigenfrequencies" (eigenfrequencies that do not converge to the partial natural frequency of the corresponding bubble) of the larger bubble do not appear; this may due to a relatively strong damping effect.^{5,6} Figure 4 shows E as a function of l , given by using different driving frequencies. In contrast with the previous example, the reversals in the cases of $\omega > \omega_{10}$ and $\omega > \omega_{20}$ are

due to the highest and second highest eigenfrequencies of the smaller bubble. The reversal in the case of $\omega < \omega_{10}$ and $\omega > \omega_{20}$ ($\omega = 0.8\omega_{10}$) is, as in the previous example, due to the second highest eigenfrequency of the smaller bubble.

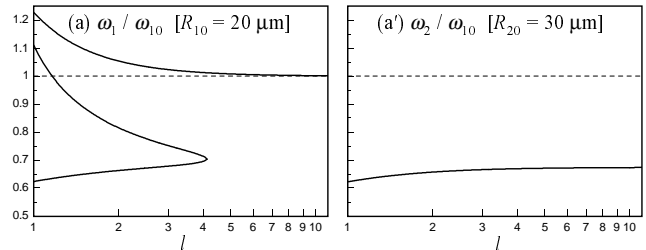


FIG. 3. Eigenfrequencies for $R_{10} = 20 \mu\text{m}$ and $R_{20} = 30 \mu\text{m}$.

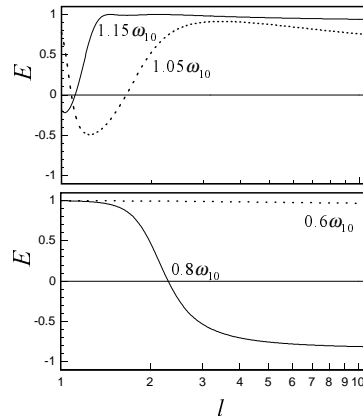


FIG. 4. $E = \cos(\phi_1 - \phi_2)$ for $\omega = 1.15\omega_{10}$, $1.05\omega_{10}$, $0.6\omega_{10}$, and $0.8\omega_{10}$.

The results given above can be summarized as follows. The first reversal, occurring when both bubbles are larger than resonance size, is due to the highest eigenfrequency of the *larger* bubble when the damping effect is sufficiently small, and to the highest eigenfrequency of the *smaller* bubble in other cases. The reversal occurring when one bubble is larger than and the other bubble is smaller than the resonance size is, in any case, due to the second highest eigenfrequency of the *smaller* bubble. These results are more complicated than the assumptions given by Doinikov and Zavtrak.^{3,4}

IV. CONCLUSION

We have discussed the influence of variations in the eigenfrequencies of gas bubbles, which result from their radiative interaction, on the secondary Bjerknes force acting between pulsating bubbles. The present theory can explain the reversal of the sign of the force in any case, and gives results that contradict the interpretations

of the reversal given by Doinikov and Zavtrak.^{3,4} Our results show that the theory given in Refs. 5 and 6 for evaluating the eigenfrequencies of interacting bubbles is a reasonable tool for accurately understanding the mechanism of the reversal. In future work⁸, the theory will be extended to systems consisting three or more bubbles, such as those discussed in Refs. 9–12.

Lastly, we remark further on the results given in Ref. 4. In that paper, the frequency of the external sound ($f = \omega/2\pi$) was assumed to be $f = 63$ kHz, which is 60 times higher than the partial resonance frequency of a bubble of $R_0 = 3$ mm (1.094 kHz); nevertheless, the reversal was observed. (In Ref. 3, the driving frequency is assumed to be comparable to the partial natural frequencies of bubbles, and the bubble radii are several tens of micrometers.) The result reveals implicitly that the mathematical model proposed in Ref. 4 predicts such a strong increase of the natural frequencies, which cannot be explained by the classical model for coupled oscillators used here. Derivation of the eigenfrequencies of Doinikov and Zavtrak’s model would be an interesting future subject.

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